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<b>(54) Title:</b> POLYOLEFIN/THERMOPLASTIC BLEND  <b>(57) Abstract</b> <p>The mechanical properties, particularly the heat distortion temperature and impact resistance, of polymeric compositions for moulding are particularly good in compositions comprising a polyolefin, an engineering thermoplastic and a compatibiliser, which compositions are capable of being moulded to give a product in which at least one of the polyolefin and engineering thermoplastic is in the form of at least some non-spherical particles dispersed in the other of the polyolefin and engineering thermoplastic.</p>		

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## POLYOLEFIN/THERMOPLASTIC BLEND

The present invention relates to a polyolefin/thermoplastic blend, in particular to a moulding composition comprising a polyolefin, an engineering thermoplastic incompatible with the polyolefin and a compatibilizer.

5 A wide variety of polymers may be moulded e.g. by injection moulding or blow moulding, to produce a range of articles including household items and industrial parts and fittings. Thus, for example, an engineering thermoplastic, e.g. nylon 6, may be moulded and the  
10 resulting article normally has good tensile strength. However, water is absorbed by the nylon and the mechanical properties of the article deteriorate with time. This deterioration takes place soon after the  
15 article is exposed to water, e.g. in a humid atmosphere, and in as little as 40 hours the properties will have settled at less favourable values. Moreover, engineering thermoplastics, e.g. nylon 6, are currently more expensive to produce than for example most  
20 polyolefins.

Polyolefins may also be moulded into articles and the resulting articles absorb much less water than e.g. nylon 6 and the properties therefore do not deteriorate significantly through water absorption. However, the  
25 mechanical properties of moulded polyolefin are worse than that of nylon 6, even when the nylon 6 has absorbed water.

Blends of a polyolefin and a thermoplastic without any additional compatibiliser may have properties worse  
30 than that of the individual polyolefin and engineering thermoplastic. To provide adhesion between the polyolefin and the thermoplastic it is known to add a compatibiliser which decreases the surface tension between the polyolefin and the thermoplastic. This may  
35 bring the strength of the blend to somewhere between

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that of the pure polyolefin and the pure thermoplastic. In such blends it is desirable to minimise the content of engineering thermoplastic whilst optimising the mechanical properties of the blend. Thus, for example, since nylon is considerably more expensive than polyolefins, reduction of nylon content of a nylon/polyolefin blend is cost advantageous, provided there is no unacceptable reduction in the physical properties of the moulded article compared with an article moulded solely from nylon or from nylon/polyolefin blends having higher nylon contents. Besides this commercial advantage, it has been established that a reduction in nylon content of a nylon/polyolefin blend leads to a reduction in water absorption, and hence to a lower water sensitivity of the blend.

In the art several attempts have been made to produce mixtures of polyolefins with other polymers to provide mixtures having properties superior to those of polyolefins. Thus, for example, EP-A-238197 describes the formation of blends of compatible polymers; however most engineering plastics are incompatible with polyolefins and this patent application does not address the problems associated with the formation of blends from incompatible polyolefin and engineering thermoplastic components.

GB-A-1596711, GB-A-1594542 and US-A-4110303 each describe blends which are typically a polyamide and a polyolefin, in the presence of a block copolymer-type compatibiliser such as SBS polymer. The blend is said to have a structure in which the polymers form interlocked network structures. The compatibiliser used is very specifically a polymer which does not include components miscible with both of the separate polymers e.g. the polyamide and polyolefin.

Thus, while one part of the compatibiliser may be miscible with, for example, a polyolefin component, the other part will be merely such that it adheres to the

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other polymer, e.g. nylon, without mixing. It is not stated that simple injection moulding will produce non-spherical particles of the disperse phase.

5 GB-A-2199584 describes blends of, for example, polypropylene and polyamide in the presence of an alkylcarboxyl-substituted polyolefin. In the extruded pipe which is prepared from the blend the polyamide is present solely as a disperse phase and it is not indicated that under conditions of low orientation, e.g. injection moulding, the particles of polyamide would be spherical; it is clear that there is no co-continuous structure, i.e. the composition is merely a dispersion of the polyamide in the polypropylene. In general, the polyamide is present at low concentrations, the preferred ratio of nylon to polyolefin being about 1:5.

15 GB-A-1081347 relates to blends of polyolefins and polyamides in which the ratio of polyamide to polyolefin is greater than 1:1. A combination of a polyamide and a molecule compatible with polyolefin is advocated as a dispersing (compatibilising) agent. This results in a dispersion of spherical particles of one polymer in the other. Similarly, GB-A-1072635 describes blends of polyamides and polyolefins in the presence of a copolymer of an alpha-olefin with an alpha, beta-unsaturated carboxylic acid in which the carboxyl groups are partially neutralised by metal ions. The product obtained appears to consist of spherical particles of one component in the other and there is no co-continuous structure.

25 EP-A-15556 describes blends of polyolefin and a second polymer in the presence of an alkylcarbonyl-substituted polyolefin as a compatibiliser. Such a blend comprises a dispersion of the second polymer in the polyolefin. It is not indicated that non-spherical particles are produced and there is no co-continuous structure. Similarly, WO 88/02764 describes blends comprising polyamides and polyolefins in the presence of

30  
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5 a compatibiliser, ethylene-vinyl acetate copolymer. The blend has a structure of particles of polyamide in a polyolefin matrix and it is not indicated that there are non-spherical particles or a co-continuous relationship between the polyolefin and the polyamide.

10 US-A-3975463 discloses blends comprising a polyolefin, a thermoplastic polymer and a saponified product of an ethylene-vinyl acetate copolymer. The moulded blend does not appear to include non-spherical particles of one component in the other and there is no co-continuous structure.

15 Important physical properties of polyolefin/engineering thermoplastic blends include the impact resistance (or notched impact strength) and the resistance of the moulded composition to distortion at high temperature. Thus distortion is measured as the heat distortion temperature (HDT), which is the temperature at which a composition distorts or deforms under a particular applied load. There has been a need  
20 to improve the physical properties, in particular the impact resistance and the HDT, of polyolefin/engineering thermoplastic blends over those blends described in the prior art.

25 The Applicant has now identified polyolefin/engineering thermoplastic blends which have improved physical properties, in particular surprisingly good impact resistance and HDT properties.

30 Generally speaking, a blend of two incompatible polymers, such as a polyolefin and a thermoplastic, will form discrete spherical particles of the one polymer dispersed in a continuous phase of the other polymer. When such blends are moulded the resulting article has the same microscopic morphology, i.e. a dispersion of one polymer in the other. However, the Applicants have  
35 discovered that blends which, on moulding, give a product in which at least one of the polyolefin and the thermoplastic is in the form, at least in part, of non-



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spherical particles dispersed in the other (including structures in which the phases are co-continuous with each other to provide extended interlocking "fingers" of each phase which can be termed particles) give  
5 compositions with unexpectedly improved physical properties without having to resort to increasing the polyamide content.

This appears in part to be associated with the use of a compatibiliser which comprises a moiety miscible in  
10 one polymeric component of the mixture and a further moiety which is either miscible with the other component or reacts therewith to add on chains of the latter which are integrated therein.

Thus, according to one aspect of the present  
15 invention, there is provided a composition comprising a multi-phase mixture of:

- (a) a polyolefin;
- (b) an engineering thermoplastic which is incompatible with (a); and
- 20 (c) a compatibiliser which decreases the surface tension between (a) and (b) and which comprises a first moiety which is miscible with one of (a) and (b) and covalently bonded thereto a second moiety which is miscible with or capable of covalently  
25 bonding to the other of (a) and (b);

said composition being capable of being injection moulded to give a product in which at least one of (a) and (b) is in the form of particles dispersed in the other of (a) and (b), at least some of said  
30 particles being non-spherical.

As indicated above, when the blends of the present invention are moulded they may produce compositions in

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which the phases of the polyolefin and the thermoplastic are co-continuous with each other. By this it is meant that neither the polyolefin nor the thermoplastic forms the dominant phase but the composition comprises  
5 particles of thermoplastic in polyolefin and particles of polyolefin in thermoplastic. The interlocking extended regions of the separate polymer phases are considered, for the purpose of the present invention, as elongated, i.e. non-spherical, particles.

10 For any particular polyolefin and any particular thermoplastic there is a "phase inversion point", at which neither polymer may be considered to be dispersed within the other. When blends are at or near the phase inversion point then, on moulding under shear or mixing  
15 under shear, they will show the above-mentioned behaviour in which one or both of the polyolefin and thermoplastic phases is/are oriented and at least some of the particles of the polyolefin or the thermoplastic are non-spherical.

20 It is particularly preferred that the particles within the composition according to the invention, after moulding, are not greater than 10 microns in their shortest diameter. This includes the extended 'fingers' of interlocking co-continuous structures, which are thus  
25 preferably not greater than 10 microns in the transverse direction. Many of the prior art compositions have produced significantly larger particles which do not, in general, provide the surprising improvement in properties associated with the compositions of the  
30 present invention.

Particles which are considered to be not substantially spherical are those which have an aspect ratio in at least one direction of greater than 1, preferably greater than 10, more preferably greater than  
35 20. As indicated above, this includes the extended interlocking regions of co-continuous mixtures.

The polyolefin and/or the engineering thermoplastic

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in the moulded article form oriented rods, elongated particles or lamellae. Elongated particles may be elongated in one or two dimensions. When the particles are elongated significantly in only one dimension they form oriented rods. When the elongation occurs significantly in two dimensions lamellae or layers are produced. This orientation does not necessarily occur uniformly throughout the sample, but preferably at least 50% by weight, more preferably at least 75% by weight, of at least one of the polymers is in the form of such lamellae, oriented rods or elongated particles. The balance of said one polymer will generally be in the form of substantially spherical particles, or at least particles with an aspect ratio considerably below 10. Most of these non-oriented particles, in a moulded article, will be found in regions where the particular moulding technique employed e.g. injection moulding, does not provide shear surfaces for the flowing polymer mixture.

The polyolefin used may be any polyolefin which is capable of being moulded. It will be recognised that such polyolefins are themselves thermoplastic, but the person of ordinary skill in the polymer blends art will readily be able to distinguish between thermoplastic polyolefins termed component (a) herein, and the thermoplastic engineering resins termed component (b) herein and discussed in more detail hereinafter. Suitable polyolefins include homo- and co-polymers of propylene, ethylene and 4-methyl-pent-1-ene. Particularly preferred is polypropylene, especially polypropylene having a melt flow rate (MFR) of from 0.3 to 15. Polypropylenes of lower MFR require less engineering thermoplastic to reach the phase inversion point of the blend, and since blends of the invention preferably have an amount of engineering thermoplastic which approaches a minimum (for cost and water absorption reasons), then polypropylenes of low MFR of

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0.3 to 8 are preferred.

The term "engineering thermoplastic" as used herein includes polyester, polycarbonate, polyether (such as polyphenyleneoxide (PPO) or polyacetal), polyamide, polystyrene, styrene-based copolymers such as high impact polystyrene (HIPS), styrene acrylonitrile (SAN), or acrylonitrile butadiene styrene (ABS), and other vinyl polymers such as polyvinylchloride (PVC).

It is preferred that the engineering thermoplastic is a polyamide, and in particular one which can be processed in the temperature range of polyolefins. Such polyamides include nylon 6, nylon 66 and polymeta-xylene adipamide (MXD6). Blends of the invention where the polyolefin is polypropylene and the engineering thermoplastic is nylon 6 are particularly preferred.

The compatibiliser is a compound which decreases the surface tension between the two (or more) polymers present in the blend, and comprises a first moiety which is miscible with one of (a) and (b) and a second moiety which is miscible with or capable of covalently bonding to the other of (a) and (b). Preferably the compatibiliser is non-ionic. The compatibiliser may preferably be a polyolefin which is attached to a component which is miscible with the thermoplastic. For example when the engineering thermoplastic is a polyamide the compatibiliser may be a polyolefin containing unsaturated carboxylic acid and/or anhydride groups. These groups may be present as a result of copolymerising the olefin with a suitable acid/anhydride-containing comonomer e.g. maleic anhydride. Alternatively the compatibiliser may be conveniently prepared by grafting an acid or anhydride onto a polyolefin.

Preferably the compatibiliser has at least one monomer in common with component (a), the polyolefin. For example, if the polyolefin is polypropylene, then the compatibiliser is preferably an acid/anhydride-

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grafted polypropylene.

Suitable carboxylic acid or anhydride groups for use in the compatibiliser include e.g. maleic acid, itaconic acid, himic acid, (5-norbornene endo 2,3 dicarboxylic acid) and their anhydrides, acrylic or methacrylic acid. In general, anhydrides are preferred to free carboxylic acids in view of their greater reactivity.

Preferably the compatibiliser contains from 0.01 to 10 wt% of the acid or anhydride-containing groups, more preferably from 0.1 to 5 wt%, especially from 0.2 to 2.5 wt%.

For the preferred component (a) polypropylene, and the preferred engineering thermoplastic nylon 6, a preferred compatibiliser which has been found to work particularly well is polypropylene on which has been grafted maleic anhydride. The amount of anhydride grafted on the polypropylene may be quite small e.g. 0.2 to 2% by weight. The less anhydride that is grafted on to the compatibiliser, the greater the minimum amount of compatibiliser required to adhere the polyolefin and engineering thermoplastic together.

The required amounts of the three essential components of the blend vary according to the particular polymers chosen. However, it is a simple procedure to determine amounts which will produce a blend which, when moulded, e.g. injection moulded, give a product according to the invention.

Once the three components have been chosen, for any particular ratio of engineering thermoplastic to compatibiliser there is only one inversion point. Thus once this ratio has been chosen the percentage of polyolefin combined with that mixture of engineering thermoplastic and compatibiliser may be altered until the phase inversion point is reached, or is so nearly approached that a moulded article formed from the blend has the necessary orientation. For example, if the

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weight ratio of compatibiliser to engineering thermoplastic is chosen to be 1:5 then a blend may be made having a weight ratio of compatibiliser: thermoplastic: polyolefin of e.g. 9:45:46. If this  
5 blend results in spherical particles of polyolefin dispersed in the engineering thermoplastic then the amount of polyolefin should be increased whilst keeping the same ratio of compatibiliser to engineering thermoplastic. For example, a blend having the ratio  
10 7:35:58 may be made. Conversely, if the initial blend results in spherical particles of engineering thermoplastic dispersed in polyolefin then the amount of engineering thermoplastic and compatibiliser should be increased, e.g. by trying a blend having the ratio  
15 11:55:34. In this way a blend can easily be obtained which is at or near the phase inversion point.

In the preferred blends of nylon 6/polypropylene (PP) /maleic anhydride grafted polypropylene (Ma-PP) typical weight percentages are 30 to 65% by weight  
20 nylon, the exact amount depending on, amongst other things, the molecular weights/viscosities of the components used.

The level of compatibiliser must be above a certain minimum. Below this minimum there is a significant  
25 decrease in nearly all the mechanical properties of the blend because the polyolefin and engineering thermoplastic are not sufficiently adhered to one another. Above the minimum level the mechanical properties increase only slightly on increasing the  
30 amount of compatibiliser.

The minimum level of compatibiliser may be determined by measuring the effect the level of compatibiliser has on physical properties, preferably the tensile strength, elongation at break and impact  
35 strength. For example, a blend may be made of a polyolefin, a compatibiliser and 20% by weight of engineering thermoplastic. This is likely to result in

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spheres of engineering thermoplastic dispersed in the blend. Increasing the proportion of the compatibiliser relative to the polyolefin, but keeping constant the percentage by weight of engineering thermoplastic in the blend, will improve the mechanical properties of the blend e.g. increase the tensile strength, elongation at break or impact strength. At the same time the size of the dispersed engineering thermoplastic spheres will decrease. The variation of tensile strength with changing levels of compatibiliser is schematically shown in Figure 1. Similar behaviour is seen for elongation at break and impact strength.

The tensile strength, elongation at break and impact strength of the blend will each level out at a (different) maximum value. The amount of compatibiliser present in the blend at the point at which all three of the tensile strength, elongation at break and impact strength have reached a maximum value is the minimum level of compatibiliser (relative to the engineering thermoplastic) necessary for optimum adhesion between the phases in the system.

It is possible to use an amount of compatibiliser which is greater than this minimum level necessary for optimum adhesion, but it is not necessary to exceed this minimum concentration to obtain the beneficial effects.

It will be appreciated that an alternative method of determining the phase inversion point is to follow the above procedure but to keep the polyolefin content fixed.

In the preferred blend of PP/nylon 6/Ma-PP the minimum amount of compatibiliser (in weight percent based on the total of the three components) is  $N/10M$  where N is the weight percent of nylon 6 and M is the weight percent of anhydride grafted on the polypropylene compatibiliser. Given the above, it is a relatively straightforward exercise to develop blends which, on moulding, will have the desired degree of orientation.

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Preferably the blends contain from 35 to 50 wt% of engineering thermoplastic, e.g. nylon 6, more preferably from 40 to 45 wt% based on (a) + (b) + (c). Once the proportion of nylon has been established, then for any given Ma content of Ma-PP, it can simply be calculated what the percentage of PP and of Ma-PP should be that which will result in a blend having the desired orientation on moulding. Thus such blends have a relatively low level of engineering thermoplastic, e.g. nylon, but because the engineering thermoplastic is oriented the blend will behave as though the thermoplastic, e.g. nylon is the continuous phase (for example with regard to its HDT and impact strength). The blend will have physical properties characteristic of e.g. nylon or high nylon blends; but will only suffer the water absorption problems associated with low nylon blends. Moreover the cost advantage of polyolefin, e.g. PP versus engineering thermoplastic, e.g. nylon means that performance approximating to engineering thermoplastic, e.g. nylon, per se can be achieved at reduced cost.

It is possible that, after injection moulding, the moulded article may have flow marks on the surface of the article. It has been found that this problem may be eliminated by using slightly higher engineering thermoplastic, e.g. nylon 6, contents or using a polyolefin, e.g. polypropylene, with a lower melt flow rate.

The composition of the present invention may advantageously additionally contain fillers such as talc and/or glass fibre. The addition of such fillers leads to a surprising improvement in modulus, tensile strength, mould shrinkage and impact strength and a reduction in water uptake, and the filled blends retain the same oriented morphology as the un-filled blend.

The composition may additionally contain rubber such as acid- or anhydride modified rubber. Such compositions are alloys with high impact strength and good tensile strength and flexural modulus.



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Surprisingly, the impact strengths of such alloys are comparable to those of so-called "super-tough nylons". Such compositions show the same oriented morphology as the non-rubber filled blends. Alternatively (a) may be an impact copolymer.

As explained hereinbefore, for a given system of fixed polyolefin type, fixed compatibiliser type, fixed thermoplastic type and fixed compatibiliser/engineering thermoplastic ratio, it is a relatively straightforward experimental procedure to determine the phase inversion point, i.e. in terms of the engineering thermoplastic concentration, of the blend. Preferably the blends of the invention, e.g. where (a) is PP, (b) is nylon 6 and (c) is Ma-PP, have an engineering thermoplastic, e.g. nylon 6, content which is no more than ten percent greater or less than the percentage of engineering thermoplastic, e.g. nylon 6, at the phase inversion point. For example if the nylon content at the phase inversion point is 45 wt%, then the nylon content of the blend is preferably in the range of 35 to 55 wt%. More preferably the variation of engineering thermoplastic content is from five percent above to five percent below the engineering thermoplastic content (wt %) at the phase inversion point.

According to a further aspect of the present invention there is provided a process for the production of a composition as hereinbefore described in which components (a), (b) and (c) are admixed.

According to another aspect of the present invention there is provided a process for producing a moulded article, said process comprising moulding a composition according to the present invention under a shear of at least  $50 \text{ sec}^{-1}$ , preferably at least  $100 \text{ sec}^{-1}$ .

The blends of the invention may be made using techniques known in the art. For example, the polymers may be dissolved in a common solvent and the solvent then removed. Another method is to melt-mix the

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polymers in a high shear mixer, such as a Banbury mixer, or other mixers such as an extruder or two-roll mixer. It is a feature of the blends of the invention that the composition (in terms of proportions and types of components) is such that, on moulding, at least one of the main phases (a) and (b) (which predominate in the blend) is in the form of particles dispersed in the other of (a) and (b), at least some of the particles being non-spherical. To achieve this morphology, it has been found to be necessary to subject the blends to shear in the moulding process. Thus a blend having a composition only slightly different from the phase inversion point for the components selected, will not exhibit the required orientation if moulded without shear, e.g. compression moulded. The amount of shear necessary to produce the desired orientation is dependent on the degree to which the composition varies from its phase inversion point. In general, a shear of at least  $50 \text{ sec}^{-1}$ , more preferably at least  $100 \text{ sec}^{-1}$  is desirable to yield the required orientation on moulding.

The composition of the present invention is particularly suitable for injection moulding directly as a dry mix. Thus, according to a further aspect of the present invention there is provided a process for producing a moulded article, said process comprising subjecting a composition according to the invention in the form of a dry mix to injection moulding.

It has been found that, generally speaking, blends that are compounded under less drastic conditions (and therefore with the lowest expense) have better colour and mechanical properties. This is very different than observed for most polymer blends where mechanical properties increase with compounding energy.

According to another aspect of the present invention there is provided a moulded article moulded from a composition according to the invention or produced by a process according to the invention.

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The following examples illustrate in a non-limiting manner the invention and its advantages over comparative examples which are indicated by the letter "C".

EXAMPLE 1: Blends 1-22

5       The components of the samples, both in terms of components and their proportions, are given in the Tables. For each sample the specified components were first dry mixed in the specified proportions, and the mix was then introduced into a Welding Engineers Inc. extruder operated at 200 RPM and having a temperature of 10       from 240°C along the barrel length. Under these conditions a melt mix was formed and extruded into an underwater pelletiser to form pellets of each sample. For each composition the pellets were dried and then 15       moulded into test pieces using an Ankerwerk injection moulder operated over the temperature range 200°C to 260°C and providing a shear on moulding of greater than 100 sec<sup>-1</sup>. The mould temperature for this operation was from 60°C to 80°C.

20       Some samples were conditioned by exposure to water, and some were dried. For each sample certain physical property measurements were performed in accordance with the following standard test procedures:

25	Tensile Strength:	ASTM D 638
	Flexural Modulus:	ASTM D 790 Method I
	Izod Notched Impact:	ASTM D 256
	Heat Distortion	
	Temperature (HDT):	ASTM D 648

30       Water absorption was measured by a simple weight gain method for a defined set of temperature, time and humidity conditions.

35       Some of the above properties are not reported for some samples, either because measurements were not attempted or because of machine failure during measurement.

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TABLE 1

	PP	PP	Nylon 6	Blend 1	Blend 2C	Blend 3
<u>Component</u>						
PP 12 (1)	100	0	0	0	30	15
PP 4 (2)	0	100	0	46	0	0
Nylon 6 (3)	0	0	100	45	50	65
Ma-PP (4)	0	0	0	9	20	20
<u>Property</u>						
Tensile Strength (MPa)	36	35	46	46	38	50
Flex. Modulus (GPa)	1.2	1.2	1.6	1.7	1.4	1.7
Notched Izod impact (J/m)	20	42	76	93	34	54
HDT (°C)	86	86	130	126	(8)	(8)
Water abs (wt %)	0	0	0.6	0.18	-	-
Morphology				COCO(6)	N-SPH(7)	COCO(6)

- (1) PP of MFR = 12  
 (2) PP of MFR = 4  
 (3) Nylon 6 of  $M_w = 30\ 000$  supplied by BASF AG (ultramid B3)  
 (4) Ma-PP of MFR = 60, grafted with 0.45wt % maleic anhydride  
 (5) After 10 days at 23°C, 50% humidity  
 (6) Cocontinuous nylon and PP phases  
 (7) Dispersed nylon 6 spheres  
 (8) HDT not measured because of machine failure

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The values in Table 1 are for samples which have been exposed to water (10 days at 23°C, 50% humidity). The blends of the invention have a lower water absorption than nylon 6, but have comparable, if not better, mechanical properties including HDT (comparable, where measured) and impact resistance (better in Blend 1). Blends 1 and 2 have very similar nylon 6 content, but blend 1 which has an oriented morphology has superior properties to blend 2.

The values in Table 2 are for dried samples. The samples all have identical levels of nylon 6 but differ in the MFR or the PP and the type of MaPP. Strength of moulded test pieces of the invention are clearly greater than those of the comparative sample on which it was possible to make measurements (Blend 7).

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TABLE 2

	Blend 4	Blend 5	Blend 6	Blend 7 C	Blend 8 C
<u>Components</u>					
PP12 (1)	0	0	0	46	0
PP 4 (2)	0	46	53	0	49
PP0.8 (9)	46	0	0	0	0
Ma-PP (a)	9	9	0	9	0
Ma-PP (b)	0	0	2	0	0
Ma-PP (c)	0	0	0	0	6
Nylon 6 (3)	45	45	45	45	45
<u>Property</u>					
HDT	126	126	123	111	111
Morphology(d)	Nyl.cont PP rods.	COCO(6) rods	PP cont. Nyl.rods	PP cont. N-SPH(7)	PP cont. N-SPH(7)
Notched Impact Strength at 20°C (J/m)	48	45	40	24	-
Notched Impact Strength at -20°C (J/m)	36	35	29	17	-

- (9) PP of MFR = 0.8  
 (a) MFR of 58 and 0.48 wt% of grafted maleic anhydride  
 (b) MFR of >1000 and 5 wt% of grafted maleic anhydride  
 (c) MFR of 19 and 0.87 wt% of grafted maleic anhydride  
 (d) Cont. = is in the continuous phase

By way of comparison it should be noted that the HDT of PP (MFR 12 or 4 or 0.8) is 75 and of Nylon 6 is 158.

TABLE 3A

BLEND OR POLYMER	NYLON (wt%)	PP MFR	[Ma] wt% grafted	Ma-PP wt%	Morphology <sup>(a)</sup>	HDdl <sup>(b)</sup>	HDcl <sup>(c)</sup>
9	40	4	0.45	8	nylrod	129	126
10	45	4	0.45	9	pprod	126	124
11	50	4	0.45	10	ppelo	126	124
12 C	55	4	0.45	11	ppsph	124	113
13 C	60	4	0.45	12	ppsph	127	109
PP4(2)	0	4		0		76	76
Nylon(3)	100			0		158	131
14	65	12	0.5	13	ppelo	136	
7 C	45	12	0.5	9	nylsph	111	
15 C	30	12	0.5	6	nylsph	112	
16 C	15	12	0.5	3	nylsph	100	
PP12(1)	0	12		0		100	
17 C	65	4		13	ppsph	130	
5	45	4	0.5	9	coco	126	
6	45	4	0.5	2	nylrod	123	
8 C	45	4	3	6	nylelo	111	
18 C	30	4	0.75	6	nylsph	105	
19 C	15	4	0.5	3	nylsph	105	
PP4(2)	0	4	0.5	0		75	
20 C	65	0.8	0.5	13	ppsph	136	
4	45	0.8	0.5	9	pprod	126	
21	30	0.8	0.5	6	nylelo	126	
22 C	15	0.8	0.5	3	nylsph	108	
PP0.8(9)	0	0.8		0		75	
Nylon(3)	100					158	

(a) rod = in rods

elo = elongated particles

sph = spherical particles

coco = cocontinuous phases

(b) HDdl: HDT at 455kPa on dried samples

(c) HDcl: HDT at 455kPa on conditioned samples (1 week/20°C/50% humidity)

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In blends 4 to 13 the tests were carried out on samples which were not ASTM standards and so the Id+20 and flexural modulus cannot be compared with those measurements made on the other blends under ASTM standard conditions. In blends 4 to 13 the testing samples had the following dimensions (in mm):

Tensile Samples: 64 x 3.3 x 2;

Impact Samples: 64 x 13 x 4;

Flexural Samples: 120 x 13 x 4;

HDT Samples: 120 x 13 x 4.

It can be seen from the tables that the HDT of blends according to the invention are higher than blends having the same engineering thermoplastic content but not having the oriented morphology. Moreover, the HDT of blends of the invention does not decrease as much with water exposure as does the HDT of blends in which the polyolefin or the engineering thermoplastic is in the form of spherical particles. It may also be seen (blends 14, 17c, 20c) that even with extremely high nylon levels (65%) there is an advantage when the blends contain non-spherical polypropylene particles rather than spherical polypropylene particles (see, for example, the Flexural Modulus values).

Figure 2 illustrates the morphology of some of the polymer blends after injection moulding. In blend 7, a comparative blend, the nylon is in the form of spherical particles dispersed in the polypropylene. Blends 4 and 10 show an oriented morphology which comprises rods of polypropylene surrounded by the nylon.

Figure 3 shows the morphology after injection moulding of blend 6. The nylon rods are seen lengthwise when viewed in side transverse section and are seen along their axes in the cross sectional view.



TABLE 3B

BLEND OR POLYMER	Id+20 (a)	strd (b)	strc (c)	flexc (d)	flexd (e)	MFR (f)
9	64	52	44	1732	1517	4
10	78	51	44	1653	1564	4.8
11	72	55	49	1909	1902	5.1
12 C	75	57	48	1789	2061	5.6
13 C	70	58	48	1674	1737	5.5
PP4(2)	42	35	31	1197	1151	4
Nylon(3)	50	73	48	1800	2650	20
14	58	61			2310	5.75
7 C	24	47		2120	2150	7.39
15 C	23	41		1770	2000	8.87
16 C	18	39		1700	1850	11.63
PP12(1)	24	39		1600	1710	12
17 C	64	61		2070	2260	4.92
5	45	50		1970	2050	4.7
6	40	50		1915	2050	4.57
8 C	49	51		2070	2180	2.23
18 C	37	43		1910	2110	4.06
19 C	27	41		1780	1970	3.88
PP4(2)	26	36		1360	1210	4
20 C	56	58		1960	2180	4.45
4	48	49		1770	1950	2.95
21	47	46		1750	1970	1.32
22 C	36	40		1600	1740	1.22
PPO.8(9)	42	37		1420	1560	0.8
Nylon(3)	36	74		2150	2580	20

(a) Id+20: Impact strength (notched Izod) on dried samples at 20°C. (J/m)

(b) strd: Tensile strength on dried samples (MPa)

(c) strc: Tensile strength on conditioned samples (MPa)

(d) flexd: Flex modulus on dried samples (MPa)

(e) flexc: Flex modulus on conditioned samples (MPa)

(f) MFR: In g/10 min at 230°C

## NOTE:

Conditioning referred to in notes (c) and (e) was the maintaining of the samples for at least one week at 20°C, 50% humidity.

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EXAMPLE 2:

Blend A was prepared having the following composition and under the following conditions:

	Blend A
5 Blended composition	
Neste VB 4006C	52.00
MXD6 (aromatic PA)	40.00
Rext 146 (Ma-g-PP)	8.00
Irganox B215	0.15
10 Dye	
<u>Compounding Conditions:</u>	
Feed rate, kg/h	10.00
Screw speed, rpm	150
Temperature	260

15 The PP/MXD6/MaPP blend was extruded in a Leistritz extruder.

20 The morphology was observed by Scanning Electron Microscope (SEM) in a backscattering mode. Extruded pellets were microtomed perpendicular to the machine direction and Ru-stained to increase phase contrast between PP and MXD6. Figure 4 shows the SEM micrograph. The morphology comprised dispersed and oriented MXD6 of particle size 10 x 200 microns.

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EXAMPLE 3: Elimination of Flow Marks

- Blend 5, although being particularly advantageous, shows deep flow marks on the surface of parts injection moulded therefrom. Substitution of Neste PPVB40 polypropylene by Neste PPVA80 polypropylene (lower MFR) or increasing the polyamide content led to elimination of the flow marks:

10	COMPOSITION					Flow Marks
	PP grade	PP wt %	Nylon 6 grade	Nylon 6 wt %	Ma-PP wt %	
	Neste PPVB40	46	Ultramid B3	45	9	Yes(=Blend 5)
	Neste PPVA80	46	Ultramid B3	45	9	No
	Exxon PP1012	42	Ultramid B3	50	8	No

15

EXAMPLE 4: Mixing Conditions

The blend containing Neste PPVA80 polypropylene exemplified in Example 2 was subjected to various compounding conditions prior to injection moulding, with the following results:

20	Compounding prior injection moulding	Colour	Tensile strength	Notched Izod Impact strength
			(MPa)*	(J/m)
	Dry blending	White	55	52
25	Single screw extrusion	light yellow	52	50
	Twin screw extrusion	dark yellow	51	47

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EXAMPLE 5: Rubber Additives

The following table shows the increase in impact strength of a 45:10 Nylon 6:MaPP composition on adding rubber or by using an impact copolymer as the polyolefin:

	Nylon 6 (Ultramid B3)	(wt %)	45	45	45
	PP (P 1012)	(wt %)	45	36	0
	PP (PD 8042)*	(wt %)	0	0	36
	MaPP (Rext 182)	(wt %)	10	10	10
10	Rubber (Exxelor VA 1803)	(wt %)	0	9	9
	Irganox B 215	(wt %)	0.15	0.15	0.15
	Yield strength	(MPa)	46	32	25
	Flexural Modulus	(GPa)	1.9	1.1	0.8
	Notched Izod	(J/m)	52	600	652
15	Impact strength at 20°C				

\* PD 8042 is an impact copolymer

EXAMPLE 6: Fillers

Various PP/PA/MaPP blends were prepared which included various quantities of fibre glass and/or talc, and various physical properties of mouldings therefrom were measured:

PP grade	Exxon PP 1012 31.5	Exxon PP 1012 36	Exxon PP 1012 31.5	Neste PP VA 040 36	Neste PP VA 040 31.5	Exxon PP 1012 70	
wt %	7	8	7	8	7	0	0
MaPP wt % (Rext 182)	7	8	7	8	7	0	0
PA 6 grade	Ultramid B3	Ultramid B3	Ultramid B3	Ultramid B3	Ultramid B3	Ultramid B3	Ultramid B3
wt %	31.5	36	31.5	36	31.5	0	70
FG grade	OCF 457 FG	OCF 457 FG	OCF 457 FG	OCF 23 EXI	OCF 23 EXI	OCF 457 FG	OCF 492 FG
wt %	30	20	20	20	30	30	30
Talc wt % (microtuff)	0	0	10	0	0	0	0
Water uptake (%)	-0.36	-	0.16	-	-	0.0	1.10
Flexural mod. (GPa)	6.8	3.3	4.9	4.9	7.4	3.2	7.1
Shrinkage Mach. (%)	0.0	-	-0.32	-	-	-0.63	0.0
Trans. (%)	+0.2	-	-0.32	-	-	-3.13	0.0
HDT at 264 (°C)	153	146	153	154	154	144	202
Tensile (MPa)	100	69	81	113	122	66	152
Izod RT (J/m)	64	44	44	99	95	48	80
- 30°C (J/m)	56	32	36	-	-	52	60
Hardness, Shore D	79	-	78	-	-	71	83

**CLAIMS:**

1. A composition comprising a multi-phase mixture of:

(a) a polyolefin;

(b) an engineering thermoplastic which is incompatible with (a); and

(c) a compatibiliser which decreases the surface tension between (a) and (b) and which comprises a first moiety which is miscible with one of (a) and (b) and covalently bonded thereto a second moiety which is miscible with or capable of covalently bonding to the other of (a) and (b);

said composition being capable of being injection moulded to give a product in which at least one of (a) and (b) is in the form of particles dispersed in the other of (a) and (b), at least some of said particles being non-spherical.

2. A composition as claimed in claim 1, wherein (c) is non-ionic.

3. A composition as claimed in any one of the preceding claims, wherein (c) and (a) have at least one monomer in common.

4. A composition as claimed in any one of the preceding claims, wherein (c) is a polyolefin to which is covalently attached a moiety which is miscible with (b) or reacts therewith.

5. A composition as claimed in any one of the preceding claims, wherein (c) is a polyolefin grafted with an unsaturated carboxylic acid and/or anhydride.

6. A composition as claimed in any one of the preceding claims, wherein (c) is maleic anhydride

grafted polypropylene.

7. A composition as claimed in any one of the preceding claims, wherein (a) is polypropylene, (b) is nylon 6, and (c) is maleic anhydride grafted polypropylene.
8. A composition as claimed in any one of the preceding claims, comprising 35 to 50% by weight based on (a) + (b) + (c) of (b).
9. A composition as claimed in any one of the preceding claims, comprising 40 to 45% by weight based on (a) + (b) + (c) of (b).
10. A composition as claimed in any one of the preceding claims, wherein the percentage by weight of (b), based on (a) + (b) + (c), is within  $\pm 5\%$  of the percentage by weight of (b) at the phase inversion point for a given ratio of (b): (c).
11. A composition as claimed in any one of the preceding claims, wherein at least 50% by weight of (a) or (b) is in the form of lamellae or elongated particles.
12. A composition as claimed in any one of the preceding claims, wherein (a) comprises a homo- or copolymer of propylene, ethylene or 4-methyl-pent-1-ene.
13. A composition as claimed in any one of the preceding claims, where (a) comprises polypropylene.
14. A composition as claimed in any one of the preceding claims, wherein (b) comprises a polyamide, polyester, polycarbonate, polyether, polystyrene, styrene-based copolymer or a vinyl polymer.
15. A composition as claimed in any one of the preceding claims, wherein (b) comprises a polyamide.
16. A composition as claimed in any one of the preceding claims, wherein (b) comprises nylon 6, nylon 66, or polymeta-xylylene adipamide (MXD 6).
17. A composition as claimed in any one of the preceding claims, wherein (b) comprises nylon 6.

18. A composition as claimed in any one of the preceding claims, including glass fibre and/or talc, or rubber.

19. A process for the production of a composition as claimed in claim 1 in which the components (a), (b) and (c) are admixed.

20. A process for producing a moulded article, said process comprising moulding a composition as claimed in any one of the preceding claims under a shear of at least  $50 \text{ sec}^{-1}$ .

21. A process as claimed in claim 20, wherein said composition is moulded under a shear of at least  $100 \text{ sec}^{-1}$ .

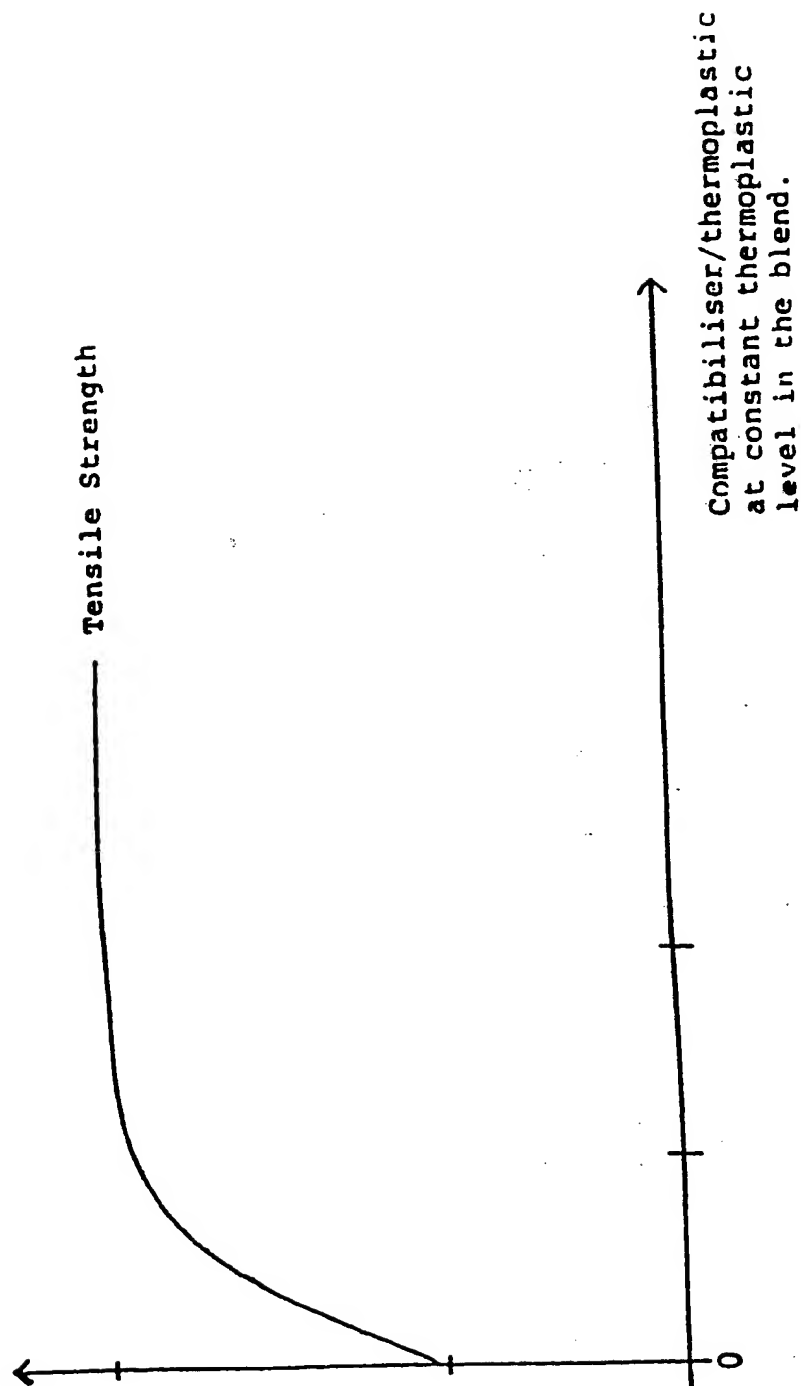
22. A process for producing a moulded article, said process comprising subjecting the components of a composition as claimed in any one of claims 1 to 18 in the form of a dry mix to injection moulding.

23. A moulded article moulded from a composition as claimed in any one of claims 1 to 18 or produced by a process as claimed in any one of claims 19 to 22.



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FIG. 1



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FIG.2C



FIG.2A

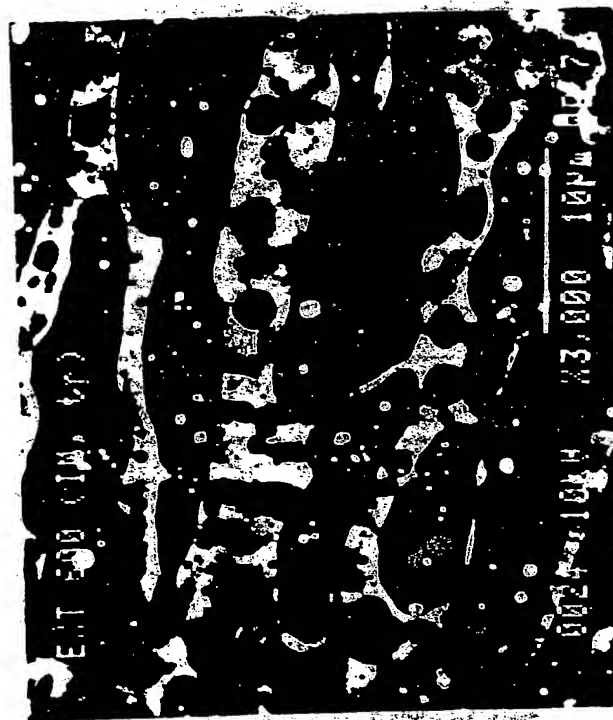


FIG.2B

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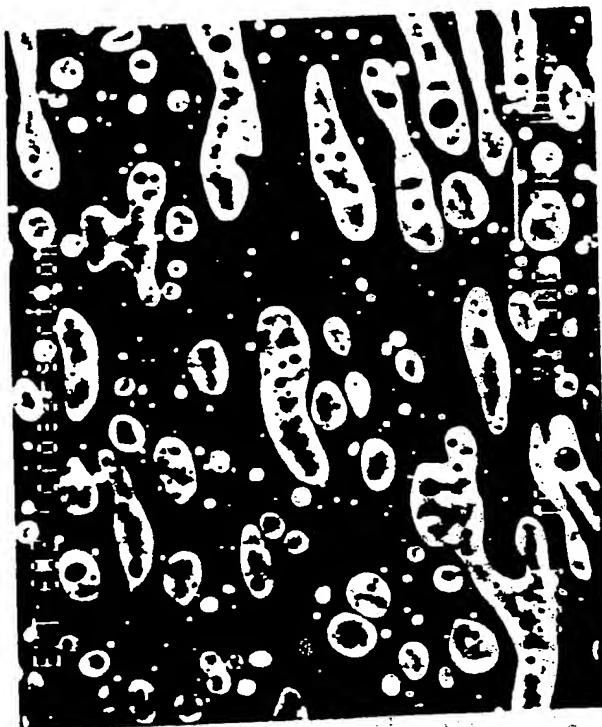


FIG.3B

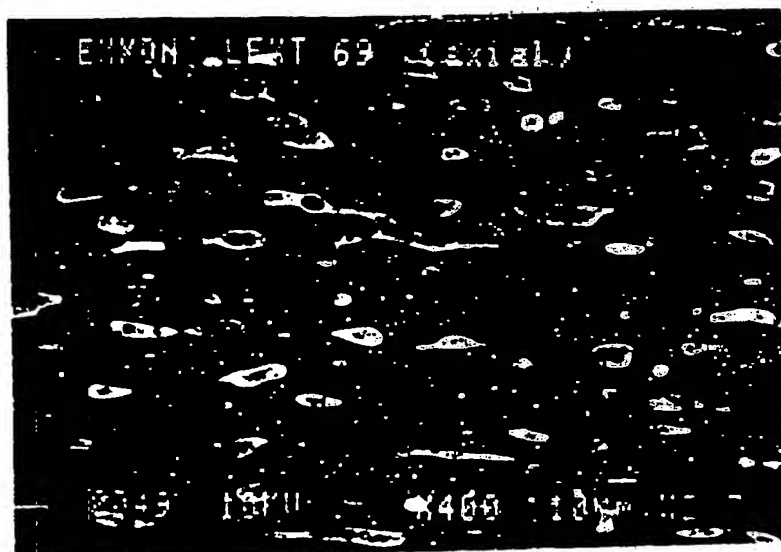


FIG.3A

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FIG. 4



**SUBSTITUTE SHEET**

# INTERNATIONAL SEARCH REPORT

International Application No PCT/US 90/05718


**I. CLASSIFICATION OF SUBJECT MATTER** (if several classification symbols apply, indicate all)<sup>6</sup>  
According to International Patent Classification (IPC) or to both National Classification and IPC  
IPC5: C 08 L 23/00, 51/00

**II. FIELDS SEARCHED**  
Minimum Documentation Searched<sup>7</sup>  
Classification System Classification Symbols  
IPC5 C 08 L  
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in Fields Searched<sup>8</sup>

III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>9</sup>		
Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
X	EP, A1, 0015556 (E.I. DU PONT DE NEMOURS AND COMPANY) 17 September 1980, see page 5, line 25 - line 30; claims 1-10 --	1-23
X	EP, A2, 0274424 (DU PONT CANADA INC.) 13 July 1988, see page 4, line 27 - line 31; abstract --	1-23
X	EP, A2, 0307802 (NIPPON PETROCHEMICALS COMPANY, LIMITED) 22 March 1989, see abstract --	1-23

- \* Special categories of cited documents:<sup>10</sup>
- "A" document defining the general state of the art which is not considered to be of particular relevance
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IV. CERTIFICATION	
Date of the Actual Completion of the International Search 18th January 1991	Date of Mailing of this International Search Report 01.02.91
International Searching Authority EUROPEAN PATENT OFFICE	Signature of Authorized Officer F.W. HECK 

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
X	EP, A1, 0331554 (ATOCHEM) 6 September 1989, see page 5, line 41 - line 47; claims 1-11  --	1-23
X	WO, A1, 8802764 (ALLIED CORPORATION) 21 April 1988, see page 6, line 27 - page 7, line 10; abstract  --  -----	1-23

**ANNEX TO THE INTERNATIONAL SEARCH REPORT  
ON INTERNATIONAL PATENT APPLICATION NO. PCT/US 90/05718**

SA 41169

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		JP-A- 55121017	17/09/80
		JP-B- 60014695	15/04/85
		US-A- 4410482	18/10/83
EP-A2- 0274424	13/07/88	GB-A- 2199584	13/07/88
		JP-A- 63252713	19/10/88
EP-A2- 0307802	22/03/89	JP-A- 2008235	11/01/90
		US-A- 4962148	09/10/90
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		JP-T- 2500983	05/04/90
		US-A- 4950515	21/08/90

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